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MICROWAVE INFLUENCES LAMINAR PREMIXED HYDROCARBON FLAMES: SPECTROSCOPIC INVESTIGATIONS

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Abstract

Low laminar burning velocities' and slow reactions propagation are among the key problem in combustion processes with low calorific gas mixtures. These mixtures have a laminar burning velocity of 10 cm/s to 15 cm/s or even below which is 37% of natural gas. Thermal use of these gases could save considerable amounts of fossil fuel and reduce CO2 emissions. Due to low burning velocities and low enthalpy of combustion, ignition and stable combustion is complex, often preventing utilization of these gases. Microwave-assisted combustion can help to solve these problems. With microwave assistance, these gas mixtures could be burned with a higher burning velocity without preheating or co-firing. Therefore, this effect could be used for flame stabilization processes in industry applications. Microwaves could also change the combustion properties, for example radical formation and flame thickness.
In this paper, we explore a possibility of using microwaves to increase the burning velocity of propane as one component in low calorific gas mixtures and also show higher productions of OH* and CH* radicals with an increase of the input microwave power. Different compositions of low calorific fuels were tested within a range of equivalence ratios from φ= 0.8 to φ= 1.3 for initial temperatures of 298 K and atmospheric conditions and microwave powers from 120 W to 600 W.

For the experiments, a standard WR340 waveguide was modified with a port for burner installation and filter elements allowing for flue gas exhaust and optical access from the side. A 2.45 GHz CW magnetron was used as microwave source, microwave measurements were carried out with a 6-port- reflectometer with integrated three-stub tuner. An axisymmetric premixed burner was designed to generate a steady conical laminar premixed flame stabilized on the outlet of a contoured nozzle under atmospheric pressure. The burner was operated with a propane mass flow of 0.2-0.4 nl/min at an equivalence ratio of φ= 0.8 to φ= 1.3. The optical techniques used in the current study are based on the flame contours detection by using the OH* chemiluminescence image technique. For every experimental case, 150 pictures were taken and averaged. Additionally, spectroscopic analysis of the flames was undertaken.

The results suggest that production of OH* radicals in the flame front increases with microwave power. For evaluation, a picture based OH* chemiluminescence and a spectrographic method were used. In addition, a 9.9% increase of the burning velocity was observed in the premixed propane-air mixture for a 66 Watt absorbed microwave power. This effect is attributed to the increased OH* (~310nm) and CH* (~420nm) radical formation, which also reduces the flame thickness. It was found that absorption of microwaves in flames is generally low, but could be improved by a customized applicator design.

1. Introduction

The field of plasma-assisted combustion aroused increased interest over the last years and new approaches for the development of combustion systems were investigated to optimize

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fuel and energy consumption. The current focus of science is on the reduction of ignition, mixing and flame stability problems and the understanding of their fundamental influences. In particular, hypersonic combustion processes with devices such as plasma burners, microwave resonators and other discharge technologies are faced with complex problems.

Previous work has shown that resonant microwaves are capable of coupling into a hydrocarbon flame front, leading to a noticeable improvement in laminar burning velocity. Furthermore, the influence of a standing wave, and therefore the field maximum of the electric field at the position of the flame have been investigated. [1, 2, 11–13] In addition to experiments with laminar flames, further studies were carried out with turbulent flames with different equivalence ratios. [11] For these conditions, microwave irradiation was studied for both continuous and pulsed. Li et al. investigated direct microwave coupling into the reaction zone, as well as the influence of plasma on the flame form. Furthermore, they found out that there was an increase in flame propagation speed due to the increase in temperature. [7] In the experiments of Shinohara, an increase of the OH intensity and burning velocity was measured, but no rotational temperature increase in the flame. Therefore, it was assumed that the change in the combustion velocity is directly related to the electron heating. [10] Further experiments focused on the strength of the electric field and the power dependence. In addition, the electron concentration and the resulting increase in reactivity of the system has been investigated experimentally and computationally with PLIF and LES. The investigations were carried out in microwave stimulation of a turbulent low-swirl flame. The experiments also focused on turbulent flames, temperature fields at ignition, burning velocity and changes in the flame position under pulsed microwave input. [6] Ju et al. made a sensitivity analysis of the free electron number for different equivalence ratios. Their results indicated that the exhaust gas could possibly absorb the majority of the microwave electron heating. [8]

2. Experimental Method

a. Laminar premixed burner setup

The burner assembly and optical measurement technology used to produce and analyze the combustion are illustrated in Fig 1. An axisymmetric premixed burner is designed to generate a steady conical laminar premixed flame stabilized on the outlet of a contoured nozzle under atmospheric pressure. The shape of the axisymmetric central contracting nozzle is designed to reduce the boundary layer thicknesses by accelerating the flow and providing a flat velocity profile at the nozzle outlet. The contoured nozzle has an outlet diameter of d1 = 10 mm and an inlet length of the premixing region of 120mm. One type K (0.5 mm) thermocouple is placed at the inlet position of the burner to monitor the temperature of combustion reactants. The current setup uses calibrated thermal mass flow controllers (MFC, EL-Flow, Bronkhorst) for controlled fuel gas supply with an uncertainty of 0.8°% of the reading value and 0.2 % of the full scale value. MFCs are controlled via software from a personal computer via Ethernet connection. Combustion air is supplied using compressed filtered air controlled by a MFC with O_2/N_2 volume ratio of 21/79. In order to create a homogenous mixture of the fuel and air a mixing chamber is used. Therefore, the temperature is controlled with a second thermocouple type K (d=0.5 mm). The outlet of the burner has been integrated into the chamber structure so that there are no irregularities between the wall and the nozzle.

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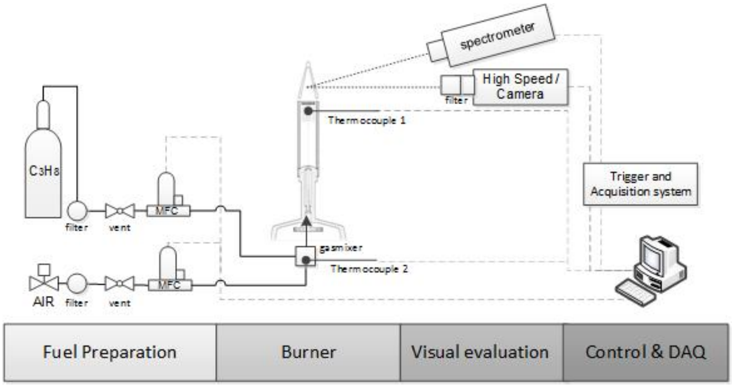

Fig. 1 Axisymmetric premixed Burner assembly at the TUBAF

b. Microwave set up

A 2.45 GHz magnetron with 1200 W nominal power was connected to a WR340 waveguide via standard launcher. A switching power supply from ALTER (CM440) was used for continuous controllable power adjustment from 120 W to 1200 W. Microwave measurements and impedance matching was carried out using a MW2104H-260EC automatic tuner from MUEGGE. The system combines an automatic impedance and power measurement system via a 6-Port-Reflectometer with a motorized three-stub tuner for load impedance matching. During the experiments, the three-stub-tuner was set to automatic impedance matching with a minimum reflection coefficient as target parameter.

For equipment safety, a circulator was installed between magnetron and tuner as well as a microwave window made from high purity fused silica between tuner and load to ensure equipment safety against flue gas condensates (water vapor).

The burner was installed in a modified WR340 waveguide build from high temperature resistant steel at quarter wavelength before the shorting wall. For exhaust gas, exit and optical access two filters with a diameter of 40 mm and length of 80 mm were integrated over the burner and lateral to the burner (see Fig. 2). The burner inlet was additionally sealed with aluminum tape for reduced microwave leakage. The waveguide and the adjacent waveguide were cooled with water-cooled aluminum cooling plates for safety. The setup was checked regularly for microwave leakage during operation via MUEGGE microwave survey meter MM3001B-110AB in order to comply with relevant health and safety norms.

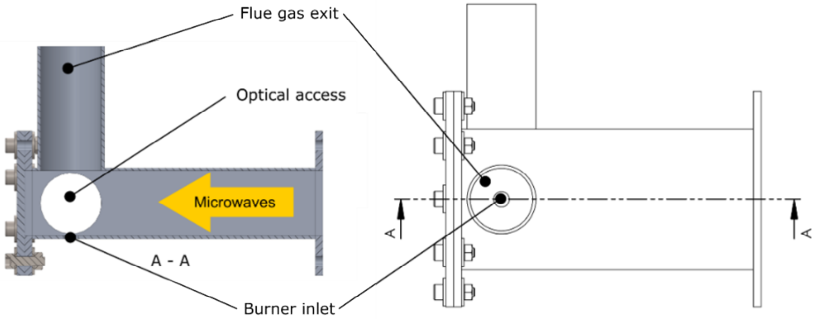

Fig. 2 Schematic representation of the microwave setup with burner installation

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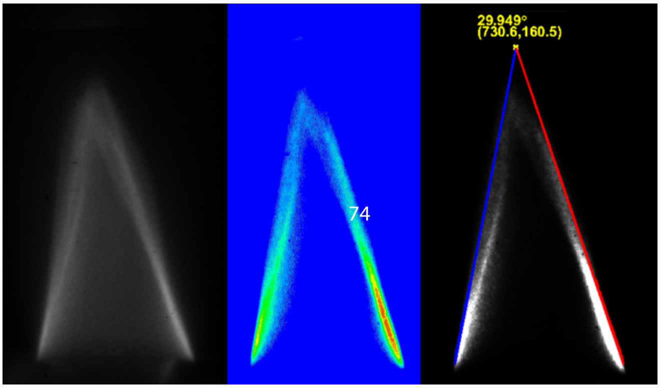

Fig. 3 The flame preparation for a picture of a propane flame with 0.3 ln/min fuel stream and at a microwave intensity of 300 W, left OH* spherical picture, middle Abel transformation and peak detection, right interpolated lines at the criteria value of 0.99 and angle calculation

c. Spectrometer and OH* chemiluminescence setup

One of the optical techniques used in the current study is based on the flame contours detection by using the OH* chemiluminescence image technique [4]. The camera used to record the OH* radical emission is an 8-bit intensified CCD camera (*mvBlueFox-224G*) with a 1600 × 1200 array. The camera is equipped with an f/3.8, f = 100 mm, achromatic UV lens (*CERCO 2073, Sodern*) combined with a short pass optical filter centered at 307 nm and having a bandwidth of ±8 nm. An Image Light Amplifier (ILA, *C9546-03L3, Hamamatsu*) is integrated in the optical system. The acquisition repetition rate of the camera is kept at 8.5 Hz. For every experimental case, 150 pictures were taken and averaged, to eliminate fluctuations. For detection of the flame angle and reaction zone, the OH* chemiluminescence image technique was used. Unlike laser-induced-fluorescence (LIF), chemiluminescence imaging is a technique, which uses the chemical excitation instead of a laser beam. The camera records the light emitted from the chemically excited OH, denoted OH*, where * represents the electronically excited state of a given radical or molecule. As in LIF imaging of OH*, a narrow optical band pass filter was placed in front of the camera lens so that the detection system only recorded light around 309±5 nm. This interference filter passed the optical emission corresponding to the $A^2\Sigma^+$–$X^2\Pi$ transition of OH. The picture preparation for the spherical OH* picture (Fig. 3), followed by a Abel transformation to a 2D distribution of the radical and a peak detection to interpolate one line for each side of the flame at the criteria of 0.99 of the maximal radical distribution in this area. The interpolated lines are used to calculate the angle between both flame fronts.

A spectrometer was used as the second examination method. The spectrometer was installed at a fixed position with a line of sight to the flame by means of a glass fiber cable. The spectrometer covers a wavelength range from 200 nm to 700 nm with a resolution of 0.2 nm. A background superposition was done to reduce background noise. This allows the detection of Signal/Noise with 300:1 and a sensitivity of 470,000 counts/µW per ms integration time.

3. Results and discussion

The experiments carried out in this thesis shall show that the laminar burning speed of propane air flames can be increased by microwave energy and that the influence on the

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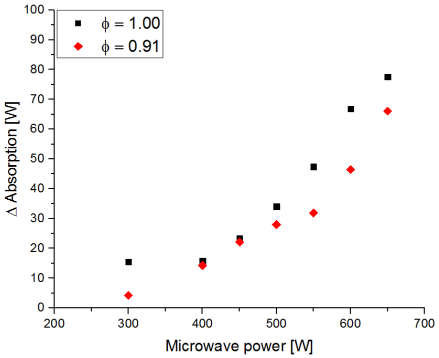

Fig. 4 Microwave absorption in a propane flame with volume flow of 0,25ln/min

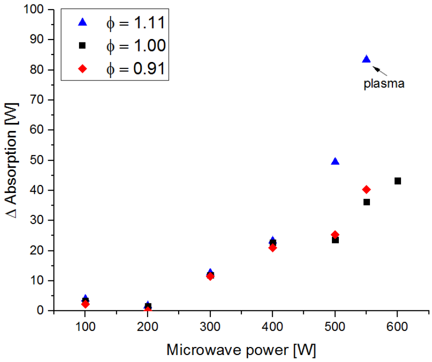

Fig. 5 Microwave absorption in a propane flame with volume flow of 0,3ln/min

radical formation in the flame front can be investigated. Results were obtained for propane-air flames at various flow conditions and equivalence ratios. The operating conditions of the volume flow rate were varied between 0.25-0.3 nl/min at fixed equivalence ratio of 0.87, 0.91, 1.0 and 1.11. For a quantification of the microwave influence, the absorption of the flame was measured for the empty WR340 waveguide and with a stabilized flame. For this procedure, the microwave power was varied. The results can be seen in Fig. 4 and Fig. 5. As in Stockman [11, 12] the absorption is a non-linear trend, this could be shown for all equivalence ratios. For the flames with a fuel rich mixture and a high fuel stream after 500 W microwave power the flame became unstable and plasma was ignited at several occasions.

Fig. 6. presents a propane flame spectrum in the wavelength between 200 nm and 700 nm for equivalence ratio Φ=0.91 and fuel stream of 0.3 nl/min. From the combustion process and the initial reactions some radicals emit light in significant wavelengths, therefore this wavelength are markers for combustion radicals such as OH* CH* and C2* which are nonstable intermediates in the combustion process.

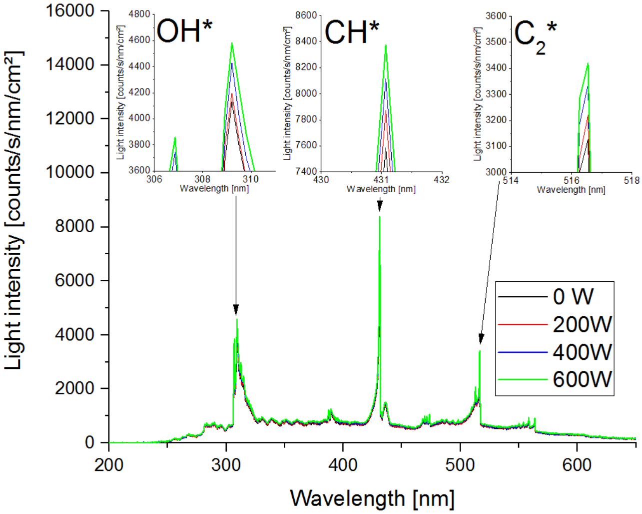

Fig. 6 Evaluation of the spectral analysis in the range from 200 nm to 700 nm at equivalence ratio of 0.91 and a fuel volume flow of 0.3 nl/min as well as the microwave influence of 0 to 600 Watt.

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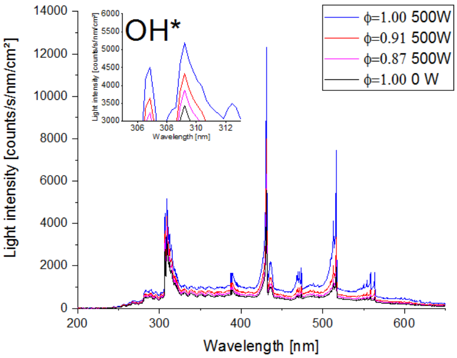

Fig. 7 Evaluation of the spectral analysis in the range from 200 nm to 700 nm at a fuel flow of 0.25 nl/min as well as the microwave influence of 0 and 500 Watt

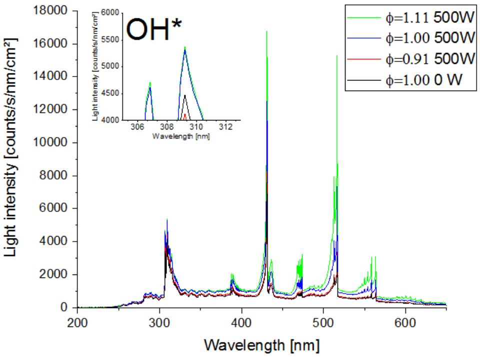

Fig. 8 Spectral analysis at a fuel flow of 0.3 nl/min as well as the microwave influence of 0 and 500 Watt for different equivalence ratios

The significant wavelengths for OH* radicals are 307 nm and 309 nm, forming a double maximum peak. They were analyzed to show the increase in the optical emission intensity when applying microwave power. OH* is typical used as a marker for the flame front, the maximal CH* (A2Δ–X2Π) emission was found to be at the wavelength 431 nm. At 516.6 nm the maximum peak for C2*(a^3 Πu–d^3Πg) radicals was found. C2* radicals can provide information about areas susceptible to soot formation. For all three investigated wavelengths it could be shown, that with an increase of microwave power, the discovered light intensity also increases at the exact same wavelength. This implies an increase in emissions in the flame front, coupled with an increase in the number of reactions in the flame. Furthermore, the influence gas mixture and volume stream was investigated. In Fig 7 the emission spectra for 200 nm to 700 nm are shown. The microwave power was fixed to 500 W and the equivalence ratio was varied. It was observed that with an increase of the equivalence ratio in the tested area the OH*, CH* and C2* radical detection increases also. Furthermore, the OH* radicals for an equivalence ratio of Φ=0.87 at 500 W are still higher than the OH* concentration at Φ=1.0 without microwave input. For the case with higher fuel stream, the situation shows the similar trend.

In Fig. 8 the result of the spectral analysis of a propane flame with a fuel volume flow of 0.3 nl/min are shown. Again, the microwave power was fixed to 500 W and the equivalence ratio was varied. It can be seen that the maximum light intensity for all wavelength increases as expected: Due to the higher amount of fuel, the reaction intensity also increases. Furthermore, it can be shown that the maximum peak value of OH* radicals rises with in the tested range. Φ=1.11 shows the highest OH* emission, while Φ=0.91 shows the lowest, the value for Φ=1.00 rises from 4440 counts/s/nm/cm² without microwave influence to 5380 counts/s/nm/cm² with influence of 500W microwave power. The same behavior can be shown for the entire considered wavelength range. The OH* concentration of Φ=1.11 is slightly higher than at Φ=1.0, but the difference is significantly lower than the difference in OH* concentrations between Φ=0.91 and Φ=1.0. This could show a similar trend to the laminar burning velocity, which also has a platoon in the area of Φ=1.0 to Φ=1.1 and decreases after the value of Φ=1.1. For this case the maximum OH* concentration at Φ=0.91 was lower than the concentration of Φ=1.0 without microwave influence. As expected, the values with higher fuel streams are for all three investigated radical cases increased.

Furthermore, with the determined flame tip angles from the OH* images the laminar burning velocity was determined.

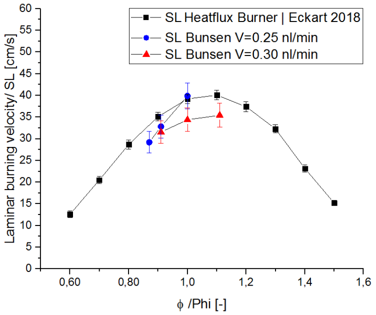

Fig. 9 Comparison of the laminar burning velocity of the cone method with the results of previous work at a Heat flux burner setup [5, 9]

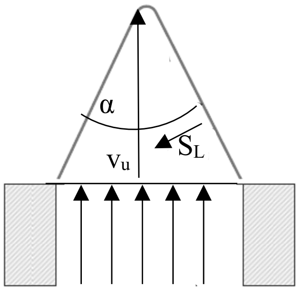

Fig. 10 Scheme for determining the laminar burning velocity (S_L) from the flame angle (α) of the cone flame and the flow velocity of the unburned fuel-air mixture (v_u)

For analysis the formula after Rallis was used [3]: $S_L - v_u \cdot \sin\left(\frac{\alpha}{2}\right)$ with flame angle (α) of the cone flame and the flow velocity of the unburned fuel-air mixture (v_u)as shown in Fig. 10. The results were compared with previous work at TUBAF with the Heatflux burner in Fig. 9. The present results for both fuel stream values are lower but show a similar trend. The accuracy of the determined burning velocity is ± 11 % and depends mainly on the uncertainty of the mass flow controller and the fluctuations of the flame tip during the experiment, resulting to an uncertainty of the flame angle. The shortened flame length, the increase of the flame tip angle, indicate the increases in the burning velocities in the flames. The shortened flame length can be interpreted as increased consumption of propane gas in the flame area closer to the burner nozzle. The burning velocity S_L was obtained experimentally by Stockman [11, 12] and Zaidi [13] with a flat flame method and pulsed microwave. In the present experiments, the cone method was used. In Fig. 11 the results for a propane flame with 3 equivalence ratio Φ=0.87, 0.91, 1.0 and a fuel stream of 0.25 nl/min are shown. It was observed that with a higher microwave, input power the flame was shortened and the illustrated flame angle increased nonlinear. Since the flame tip fluctuated during the averaging of 150 images, this has an effect on the error bars. Thus, in this case, the error detected does not provide an accurate interpretation of the trend. In Fig. 12 the volume flow was increased to 0.3 nl/min. Because of the higher unburned gas mixture speed at the nozzle the angle of the flame decreases without microwave input.

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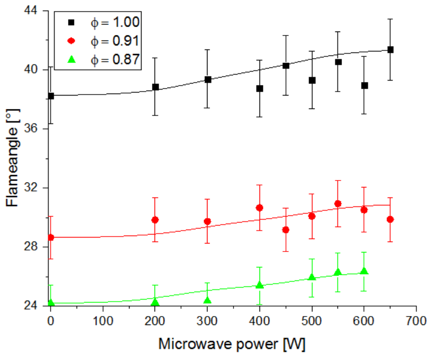

Fig. 11 Evaluation of the determined flame angles at equivalence ratios of Φ=0.87 to 1.0 and a fuel flow of 0.25 nl/min

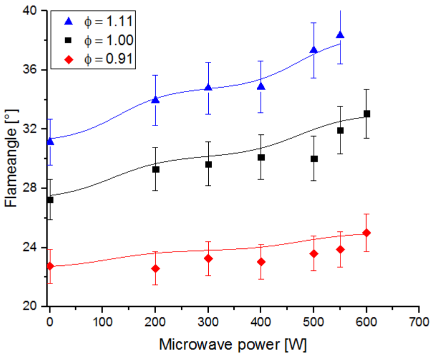

Fig. 12 Evaluation of the determined flame angles at equivalence ratios of Φ=0.91 to 1.11 and a fuel flow of 0.3 nl/min

After this, the input microwave power parameter was set as before. In this second case the influence of the microwave and the trend seems to be more clear, especially for the fuel rich conditions at Φ=1.11. With the increase of the microwave power, the flame angle rises for all three equivalence ratios. Higher equivalence ratios in the fuel rich case could be of further interest in the future.

4. Conclusions & Outlook

We observed the enhancements of the burning velocity in a premixed propane-air flame by irradiating microwave power in a wide range of equivalence ratios at atmospheric conditions. In addition, the intensity of the microwave power has a significant influence on the optical emission intensity of OH* radicals. This has been thoroughly investigated by spectrometer and OH* image investigations. Therefore, it was shown that microwave energy couples in the reaction front of a premixed propane flame and significantly influences the inter reaction chains. From this can be concluded that the emission behavior of the flame will also be influenced. For that reason, further investigations on the reaction paths will follow. The measurement of the temperature distribution inside the flame zone, the estimation of the electron energy distribution function and the measurements of radical densities will be helpful for understanding the mechanism of burning velocity enhancement. In addition, the effects of microwave input on the exhaust gas composition are of further interest.

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